

(12) INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(19) World Intellectual Property
Organization
International Bureau



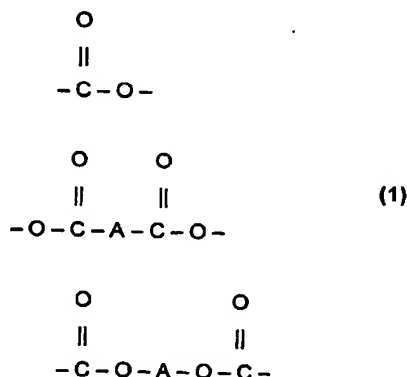
(43) International Publication Date
15 September 2005 (15.09.2005)

PCT

(10) International Publication Number
WO 2005/085340 A1

- (51) International Patent Classification⁷: **C08K 5/101**
- (21) International Application Number: **PCT/GB2005/000805**
- (22) International Filing Date: **2 March 2005 (02.03.2005)**
- (25) Filing Language: **English**
- (26) Publication Language: **English**
- (30) Priority Data:
0404620.7 **2 March 2004 (02.03.2004)** **GB**
- (71) Applicant (for all designated States except US): **CRODA INTERNATIONAL PLC [GB/GB]; Cowick Hall, Snaith, Goole, North Humberside DN14 9AA (GB).**
- (72) Inventors; and
- (75) Inventors/Applicants (for US only): **PARKER, David, Andrew [GB/GB]; c/o Croda International plc, Cowick Hall, Snaith, Goole, North Humberside DN14 9AA (GB). MALTBY, Adam [GB/GB]; c/o Croda International plc, Cowick Hall, Snaith, Goole, North Humberside DN14 9AA (GB). READ, Martin [—/GB]; c/o Croda International plc, Cowick Hall, Snaith, Goole, North Humberside DN14 9AA (GB). McCOY, Philip [—/GB]; c/o Croda International plc, Cowick Hall, Snaith, Goole, North Humberside DN14 9AA (GB).**
- (74) Agent: **COATES, Ian, Harold; Marks & Clerk, 45 Grosvenor Road, St Albans AL1 3AW (GB).**
- (81) Designated States (unless otherwise indicated, for every kind of national protection available): **AE, AG, AL, AM, AT, AU, AZ, BA, BB, BG, BR, BW, BY, BZ, CA, CH, CN, CO, CR, CU, CZ, DE, DK, DM, DZ, EC, EE, EG, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MA, MD, MG, MK, MN, MW, MX, MZ, NA, NI, NO, NZ, OM, PG, PH, PL, PT, RO, RU, SC, SD, SE, SG, SK, SL, SM, SY, TJ, TM, TN, TR, TT, TZ, UA, UG, US, UZ, VC, VN, YU, ZA, ZM, ZW.**
- (84) Designated States (unless otherwise indicated, for every kind of regional protection available): **ARIPO (BW, GH, GM, KE, LS, MW, MZ, NA, SD, SL, SZ, TZ, UG, ZM, ZW), Eurasian (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM), European (AT, BE, BG, CH, CY, CZ, DE, DK, EE, ES, FI, FR, GB, GR, HU, IE, IS, IT, LT, LU, MC, NL, PL, PT, RO, SE, SI, SK, TR), OAPI (BF, BJ, CF, CG, CI, CM, GA, GN, GQ, GW, ML, MR, NE, SN, TD, TG).**
- Published:**
- with international search report
 - before the expiration of the time limit for amending the claims and to be republished in the event of receipt of amendments
- For two-letter codes and other abbreviations, refer to the "Guidance Notes on Codes and Abbreviations" appearing at the beginning of each regular issue of the PCT Gazette.*

(54) Title: **ALIPHATIC ESTER COMPOUNDS AS SLIP AGENTS IN POLYESTER POLYMERS**



(57) Abstract: Use of a compound of general formula (1) as a slip agent in a PET polymer, wherein: R and R¹ represent hydrocarbon moieties, each hydrocarbon moiety comprising 6 to 24 carbon atoms and wherein R and/or R¹ may be linear, branched chain, saturated or contain one or more double bonds; X represents one of the moieties (a), wherein A represents a hydrocarbon moiety comprising 2 to 36 carbon atoms and may be linear, branched chain, saturated or contain one or more double bonds.

ALIPHATIC ESTER COMPOUNDS AS SLIP AGENTS IN POLYESTER POLYMERS

The present invention relates to polymer additives. It is particularly applicable to additives which reduce the coefficient of friction of a polyester polymer, such as PET.

Poly(ethylene terephthalate) (PET) is an important plastics material, widely used in the manufacture of moulded polyester articles and films. The key advantages of using PET are:

10

- High clarity
- Light weight
- Good processability
- Excellent barrier properties against oxygen and carbon dioxide
- 15 • Good impact resistance
- Tough – virtually unbreakable
- Economic

Largely as a consequence of the above properties, the most important plastic application for PET homopolymer and copolymers is in the manufacture of bottles.

PET bottles are produced predominantly using a two stage stretch blow moulding process. Firstly a preform is produced by injection moulding. This is a relatively thick – walled part with the neck features moulded during this process. The preform is then reheated in a reheat blow machine which stretches the preform by a stretching pin and inflates it by blowing air into the mould to give the desired shape. This gives a biaxially orientated container which provides improved properties such as clarity and gas barrier performance. This is especially important for carbonated drink containers.

PET bottles may also be manufactured by injection blow moulding which is a 2-stage technique performed on a single machine. The preform is injection moulded and whilst still hot is moved to a blowing station where it is blown up to the desired shape. This is the preferred technique for small containers requiring specific neck detail or finish and produces containers that are less biaxially orientated.

A major difficulty in fabricating articles from PET is the relatively high coefficient of friction of the polymer. In the manufacture of bottles this problem can manifest itself in a number of ways:

5

- Less than optimum packing density when performs are packed into a box with concomitant higher storage and transportation costs.
- Poor flow on conveying equipment and hence reduced throughput
- Surface defects due to poor scratch resistance

10

There is thus a need for an effective additive system for PET which reduces the coefficient of friction of the polymer and thus overcomes the above deficiencies.

15

Additives that are effective in reducing the coefficient of friction of polymers are known in the industry as slip additives. However, in order to be acceptable for beverage containers, the fabricated PET bottle must exhibit low colour and high clarity, with low taste and odour and be non-toxic. This imposes other important requirements on a slip agent in addition to its friction-reducing properties.

20

The conventional slip agents of choice in the plastics industry are fatty amides. These additives are widely used in polyolefins such as polyethylene, polypropylene, and related copolymers. Fatty amides employed as slip additives are generally manufactured from fatty acids containing between 16 and 22 carbon atoms and are characterised by a variety of structural forms:

25

- Primary amides which can be either monounsaturated (as exemplified by erucamide and oleamide) or saturated (as exemplified by stearamide and behenamide)
- Secondary amides as exemplified by stearyl erucamide and oleyl palmitamide
- Bis amides such as ethylene bis stearamide

30

In view of their widespread usage in polymer systems, it might appear logical to consider fatty amides as slip agents for polyesters such PET. However we have established that although fatty amides do demonstrate some friction reducing properties in PET, the lowering of the coefficient of friction is much less than in

35

polyolefins. Moreover all amides cause discolouration in injection moulded PET which will severely restrict their utility in this polymer.

Those skilled in the art will be aware that separate and different classes of polymers have widely different chemical compositions and different molecular architectures. Thus, polyester polymers such as PET cannot be compared with polyvinyl chloride (PVC), polyamides such as nylon, or other classes of polymer. Not only do they behave differently as polymers, but different slip agents are required with different polymer classes. That is to say, one cannot extrapolate or predict how a particular compound, or mixture of compounds, will perform as slip agents in one agent based on its performance as a slip agent in a different class of polymers.

Conventional slip agent technology cannot therefore be readily applied to in PET. This is particularly the case in bottle (preform) manufacture where in addition to low coefficient of friction, other strict requirements with regard to colour, taste and odour must be met.

It is therefore an object of the present invention to provide compositions having improved slip and anti-block characteristics when used in polymers such as PET and wherein other properties of the polymer are not adversely affected.

According to a first aspect of the present invention there is provided use of a compound of general Formula 1 as a slip agent in a polyester polymer:

25



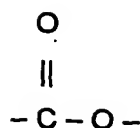
wherein: R and R¹ represent hydrocarbon moieties, each hydrocarbon moiety comprising 1 to 34 carbon atoms and wherein R and/or R¹ may be linear, branched chain, saturated or contain one or more double bonds; and wherein

30

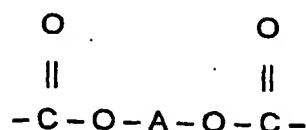
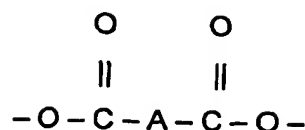
4

X represents one of the moieties:

5



10



- 15 wherein A represents a hydrocarbon moiety comprising 2 to 36 carbon atoms and may be linear, branched chain, saturated or contain one or more double bonds.

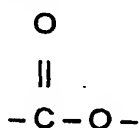
Preferably the total number of carbon atoms in R, R¹ and X is greater than 16 and more preferably greater than 22.

20

In a particularly preferred embodiment the total number of carbon atoms in R, R¹ and X is greater than 35.

Preferably X represents the moiety

25



and the total number of carbon atoms in R, R¹ and X is between 23 and 44.

- 30 In a particularly preferred embodiment the composition of general Formula 1 is selected from the group comprising stearyl stearate, stearyl behenate, behenyl behenate, ethylene glycol distearate, ethyl behenate, behenyl acetate, palmityl myristate, palmityl palmate or mixtures thereof.

- 35 In a particularly preferred embodiment the polyester polymer is selected from the group comprising:-

- poly(butylenes terephthalate)
 poly(cyclohexanedimethylene terephthalate)
 poly(ethylene isophthalate)
 5 poly(ethylene 2,6-naphthalenedicarboxylate)
 poly(ethylene phthalate)
 poly(ethylene terephthalate).

10 Preferably said composition of general Formula 1 is present in said polymer in an amount of between 0.1% to 1.0% wt/wt.

In a particularly preferred embodiment said composition is present in said polymer in an amount of between 0.2% to 0.75% wt/wt.

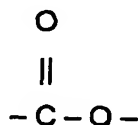
- 15 According to a second aspect of the invention there is provided a polyester polymer incorporating one or more slip agents of general Formula 1:



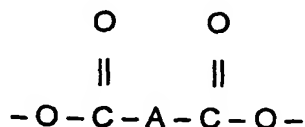
- 20 wherein: R and R¹ represent hydrocarbon moieties, each hydrocarbon moiety comprising 1 to 34 carbon atoms and wherein R and/or R¹ may be linear, branched chain, saturated or contain one or more double bonds; and wherein

X represents one of the moieties:

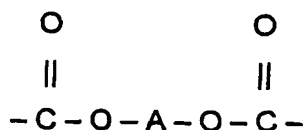
25



30



35



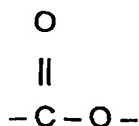
wherein A represents a hydrocarbon moiety comprising 2 to 36 carbon atoms and may be linear, branched chain, saturated or contain one or more double bonds.

- 5 Preferably the total number of carbon atoms in R, R¹ and X is greater than 16 and more preferably greater than 22.

In a particularly preferred embodiment the total number of carbon atoms in R, R¹ and X is greater than 35.

10

Preferably X represents the moiety



15

and the total number of carbon atoms in R, R¹ and X is between 23 and 44.

- In a particularly preferred embodiment the slip agent of general Formula 1 is selected from the group comprising stearyl stearate, stearyl behenate, behenyl behenate, ethylene glycol distearate, ethyl behenate, behenyl acetate, palmityl myristate, palmityl palmate or mixtures thereof.
- 20

- Where the polymer is intended for fibre production the slip agent is preferably not a stearyl ester such as stearyl stearate or other additives specifically named in GB2152061 (Snia Fibre SpA). The additives referred to in GB2152061 are described in the context of extruding fibres, not in the context of a slip additive, or in the context of preforms or bottles as in the present application.
- 25

Preferably said polymer is selected from a group comprising:-

- poly(butylenes terephthalate)
- poly(cyclohexanedimethylene terephthalate)
- 5 poly(ethylene isophthalate)
- poly(ethylene 2,6-naphthalenedicarboxylate)
- poly(ethylene phthalate)
- poly(ethylene terephthalate)
- and co-polymers thereof.

10

Preferably said slip agent(s) are present in said polyester polymer in an amount of between 0.1% to 1.0% wt/wt.

- 15 In a particularly preferred embodiment said slip agent(s) are present in said polyesterpolymer in an amount of between 0.2% to 0.75% wt/wt.

According to a third aspect of the present invention there is provided a method of treating a polymer to increase the slip of said polymer said method comprising incorporating into said polymer a composition of general Formula 1 as defined above.

20

Preferably said polymer is selected from a group comprising:-

- poly(butylenes terephthalate)
- 25 poly(cyclohexanedimethylene terephthalate)
- poly(ethylene isophthalate)
- poly(ethylene 2,6-naphthalenedicarboxylate)
- poly(ethylene phthalate)
- poly(ethylene terephthalate)
- 30 and co-polymers thereof.

Preferably the said composition of general Formula 1 is present in said polymer in an amount of between 0.1% to 1.0% wt/wt.

- 35 In a particularly preferred embodiment said composition of general Formula 1 is present in said polymer in an amount of between 0.2% to 0.75% wt/wt.

According to further aspects of the present invention there is provided a pre-form and a container made from a polymer as described herein, incorporating a slip agent of general Formula 1.

5

Preferably said container is formed from a polymer selected from a group comprising:-

- poly(butylenes terephthalate)
- 10 poly(cyclohexanedimethylene terephthalate)
- poly(ethylene isophthalate)
- poly(ethylene 2,6-naphthalenedicarboxylate)
- poly(ethylene phthalate)
- poly(ethylene terephthalate)
- 15 and co-polymers thereof.

According to a still further aspect of the present invention there is provided a film made from a polyester polymer as described herein incorporating a slip agent of general Formula 1.

20

Preferably said film is formed from a polymer selected from a group comprising:-

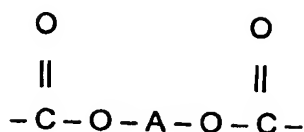
- poly(butylenes terephthalate)
- poly(cyclohexanedimethylene terephthalate)
- 25 poly(ethylene isophthalate)
- poly(ethylene 2,6-naphthalenedicarboxylate)
- poly(ethylene phthalate)
- poly(ethylene terephthalate)
- and co-polymers thereof.

30

The present invention also extends to include a composition comprising a copolymer of a polyester and a compound of general Formula 1 wherein: R and R¹ represent hydrocarbon moieties, each hydrocarbon moiety comprising 1 to 34 carbon atoms and R and/or R¹ may be linear, branched chain, saturated or contain
35 one or more double bonds;

9

X represents one of the moieties:



- 15 wherein A represents a hydrocarbon moiety comprising 2 to 36 carbon atoms and may be linear, branched chain, saturated or contain one or more double bonds.

20 The present invention therefore relates to the discovery of a novel range of slip additives for polyester polymers such as PET which are highly effective in lowering the coefficient of friction of the fabricated article whilst maintaining low colour and high clarity. More particularly, additives conforming to this invention afford a rapid reduction in the coefficient of friction that is maintained during long-term storage of the moulded part. This is particularly critical in the production of preforms and bottles from PET.

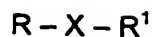
25

30 The term "PET" as used herein in describing this invention has a broad meaning. It includes all polymeric and copolymeric forms of poly (ethyleneterephthalate). The compounds of this invention are also effective slip agents for other polyester polymers and copolymers as exemplified by polybutylene terephthalate and poly (ethylene naphthalate). Thus the term PET should be considered, in this context, to be a generic term to include all polymers derived from aromatic diacids including all terephthalate polymers and their derivatives, both known and those yet to be discovered.

35

The additives of this invention conform to the general structure:

10



where R and R' are hydrocarbon moieties, each comprising 1 to 34 carbon atoms, and may be linear or branched chain, and may be fully saturated or contain one or
5 more double bonds.

X conforms to one of the following structures:



10

or



15

or



where A is a hydrocarbon moiety comprising 2 to 36 carbon atoms, and may be
20 linear or branched and be fully saturated or contain one or more double bonds.

In a preferred embodiment of the invention the total number of carbon atoms contained within R, R' and X is greater than 22 and preferably greater than 35.

25 Examples of preferred additives conforming to this invention are stearyl stearate, stearyl behenate, behenyl behenate, ethylene glycol distearate, ethyl behenate, behenyl acetate, palmityl myristate, palmityl palmate or mixtures thereof.

To achieve the required level of slip performance in PET, the additives of
30 this invention are incorporated at levels of between 0.1% and 1% and preferably between 0.2% and 0.75% wt/wt.

The slip additives of this invention may be incorporated into the polymer by a number of processes well known to those skilled in the art. For example they may
35 be added directly to the resin by melt dosing at the point of extrusion, by conventional masterbatch addition or by incorporation using liquid colour systems.

Examples

To demonstrate the effectiveness of the aforementioned additives in
5 reducing the friction of PET surfaces the following procedure was adopted.

A PET co-polymer (IV 0.8) suitable for the manufacture of bottles and other
food packaging containers by injection moulding, blow moulding or a combination of
both was used. The PET was dried for 8 hours at 145°C and the additive coated
10 directly onto the surface of the polymer by tumble mixing whilst the polymer was still
hot.

The PET was moulded into 100 x 50 x 2 mm plaques on a 35 tonne lock
injection-moulding machine using the following conditions:

15

Temperature:	All zones at 270°C
Injection Pressure:	85 Bar
Shotsize:	29.0mm
Pack:	20 Bar; 3 secs
20 Hold:	75 Bar; 3 secs
Cooling:	20 secs
Tool Temp:	10°C

The Coefficient of Friction (static and kinetic) of the resulting plaques were
25 then measured on a Lloyd LRX tensile tester and a 10N load cell at the following
time intervals after moulding – 1 hour, 24 hours, 1 week and 2 weeks. The friction
method was adapted from ASTM 1894. The sledge weight including the plaque was
1000g and the area of surface contact between the two plaques was 50mm x 50mm
(see diagram). The test was run over a distance of 60mm at 150mm/min. Each test
30 was conducted 5 times for each time interval using new plaques on each run. A
diagram of the test apparatus is shown in Figure 1.

Due to the nature of PET the friction can vary from day to day depending on
process and ambient conditions and its hygroscopic nature. The coefficient of
35 friction recorded for PET with no additives was generally between 0.5 and 1.2. To
enable comparisons to be made for experiments carried out on different days blank

runs were carried out before and after each series of PET + additives on each day. The results are reported as a percentage of the blank as it was observed that a given additive would give a proportionally lower result on a day where a low friction was recorded for the blank.

5

A number of additives of mixed ester composition were also tested and representative compositions are given in Table II labelled Formulations 1 – 6.

A summary of the slip additives tested is given below:

TABLE 1

Sample	Conc %	Coefficient of Friction (% of blank)				
		Static			Dynamic	
		initial	1 day	7 days	initial	1 day
erucamide	0.5	85	NR	76	78	NR
behenamide	0.5	62	NR	100	71	NR
GMS 90	0.5	0	NR	109	0	NR
GMB	0.5	0	NR	124	0	NR
PEG200 dierucate	0.5	89	NR	140	72	NR
PEG200 dioleate	0.5	NR	fail	NR	NR	fail
Pentaerythritol dioleate	0.5	NR	fail	fail	NR	fail
Pentaerythritol monooleate	0.5	NR	fail	fail	NR	fail
PEG400 monolaurate	0.5	NR	fail	fail	NR	fail
sorbitan monostearate	0.5	NR	fail	fail	NR	fail
Calcium stearate	0.5	NR	NR	124	NR	NR
pentaerythritol tetrastearate	0.5	95	NR	86	107	NR
butyl stearate	0.5	86	NR	87	70	NR
EthylHexyl stearate	0.5	80	NR	83	62	NR
lauryl palmitate	0.5	74	NR	85	63	NR
oleyl behenate	0.5	122	NR	173	80	NR
behenyl behenate	0.5	98	NR	94	118	NR
lauryl behenate	0.5	55	NR	55	62	NR
lauryl behenate	0.2	60	NR	72	57	NR
oleyl erucate	0.5	123	NR	fail	114	fail
EGDS	0.5	92	90	68	86	95
cetostearyl phthalate	0.5	45	83	82	54	95
butyl behenate	0.05	49	83	34	57	64
butyl behenate	0.1	33	65	33	52	62
butyl behenate	0.2	18	38	24	42	55
butyl behenate	0.5	15	30	16	27	33
ester Formulation 1	0.1	64	50	58	91	59
ester Formulation 1	0.2	38	38	36	45	38
ester Formulation 1	0.3	29	34	30	32	30
ester Formulation 2	0.2	69	51		80	62
ester Formulation 3	0.2	67	54		64	44
stearyl stearate	0.1	100	92	77	131	92
stearyl stearate	0.2	34	41	37	41	39
stearyl stearate	0.3	33	40	33	32	36
stearyl behenate	0.1	60	70	58	135	122
stearyl behenate	0.2	33	41	91	43	44
stearyl behenate	0.3	42	49	53	57	48

Sample	Conc %	Coefficient of Friction (% of blank)					
		Static			Dynamic		
		initial	1 day	7 days	initial	1 day	7
stearyl palmitate	0.1	175	109	312	270	162	
stearyl palmitate	0.2	62	49	89	69	54	
stearyl palmitate	0.3	33	39	30	43	36	
ester Formulation 4	0.2	48	46	48	56	48	
ester Formulation 5	0.2	68	57	74	72	55	
ester Formulation 6	0.2	58	46	72	85	63	
Ethyl behenate	0.2	42	49	59	33	42	
Behenyl acetate	0.2	34	42	54	31	34	
Dilauryl succinate	0.2	74	69		68	59	

TABLE 2**Formulation 1**

		alcohol				
		lauryl	myristyl	palmityl	stearyl	arachidyl
acid	laurate	<1	<1	<1	<1	<1
	myristate	<1	14-17	8-12	4-6	<1
	palmitate	<1	32-38	20-24	8-12	<1
	stearate	<1	<1	<1	<1	<1

Formulation 2

		alcohol				
		lauryl	myristyl	palmityl	stearyl	arachidyl
acid	laurate	<1	<1	<1	<1	<1
	myristate	<1	13-16	30-34	12-14	<1
	palmitate	<1	8-10	18-22	7-10	<1
	stearate	<1	<1	<1	<1	<1

Formulation 3

		alcohol				
		lauryl	myristyl	palmityl	stearyl	arachidyl
acid	laurate	<1	<1	<1	<1	<1
	myristate	<1	<1	18-22	9-11	<1
	palmitate	<1	0.5-1.5	41-45	20-24	<1
	stearate	<1	<1	<1	<1	<1

Formulation 4

		alcohol				
		lauryl	myristyl	palmityl	stearyl	arachidyl
acid	laurate	<1	<1	<1	<1	<1
	myristate	<1	7-9	4-6	2-4	<1
	palmitate	<1	16-19	10-12	5-7	<1
	stearate	<1	<1	2-4	40-45	<1

TABLE 2 continued**Formulation 5**

		alcohol				
		lauryl	myristyl	palmityl	stearyl	arachidyl
acid	laurate	<1	<1	<1	<1	<1
	myristate	<1	7-9	4-6	2-4	<1
	palmitate	<1	16-19	10-12	4-6	<1
	stearate	<1	<1	<1	<2	<1
	arachidate	<1	<1	<1	1-3	<1
	behenate	<1	<1	<1	40-45	<1

Formulation 6

		alcohol				
		lauryl	myristyl	palmityl	stearyl	arachidyl
acid	laurate	<1	<1	<1	<1	<1
	myristate	<1	7-9	4-6	2-4	<1
	palmitate	<1	16-19	10-12	48-53	<1
	stearate	<1	<1	<1	<1	<1

TABLE 2 continued

Formulation 1

		alcohol				
		lauryl	myristyl	palmityl	stearyl	arachidyl
acid	laurate	<1	<1	<1	<1	<1
	myristate	<1	14-17	8-12	4-6	<1
	palmitate	<1	32-38	20-24	8-12	<1
	stearate	<1	<1	<1	<1	<1

Formulation 2

		alcohol				
		lauryl	myristyl	palmityl	stearyl	arachidyl
acid	laurate	<1	<1	<1	<1	<1
	myristate	<1	13-16	30-34	12-14	<1
	palmitate	<1	8-10	18-22	7-10	<1
	stearate	<1	<1	<1	<1	<1

Formulation 3

		alcohol				
		lauryl	myristyl	palmityl	stearyl	arachidyl
acid	laurate	<1	<1	<1	<1	<1
	myristate	<1	<1	18-22	9-11	<1
	palmitate	<1	0.5-1.5	41-45	20-24	<1
	stearate	<1	<1	<1	<1	<1

5

Formulation 4

		alcohol				
		lauryl	myristyl	palmityl	stearyl	arachidyl
acid	laurate	<1	<1	<1	<1	<1
	myristate	<1	7-9	4-6	2-4	<1
	palmitate	<1	16-19	10-12	5-7	<1
	stearate	<1	<1	2-4	40-45	<1

TABLE 2 continued

Formulation 5

		alcohol				
		lauryl	myristyl	palmityl	stearyl	arachidyl
acid	laurate	<1	<1	<1	<1	<1
	myristate	<1	7-9	4-6	2-4	<1
	palmitate	<1	16-19	10-12	4-6	<1
	stearate	<1	<1	<1	<2	<1
	arachidate	<1	<1	<1	1-3	<1
	behenate	<1	<1	<1	40-45	<1

Formulation 6

		alcohol				
		lauryl	myristyl	palmityl	stearyl	arachidyl
acid	laurate	<1	<1	<1	<1	<1
	myristate	<1	7-9	4-6	2-4	<1
	palmitate	<1	16-19	10-12	48-53	<1
	stearate	<1	<1	<1	<1	<1

- 5 The superior performance of the additives of this invention can be readily appreciated by reference to the above results.

Conventional amide slip agents, as exemplified by erucamide (4), lowered the coefficient of friction to between 62% and 71% of the blank but resulted in severe yellowing of the polymer. In addition, the effect was short lived and after 7 days was back to the control value.

Additives conforming to this invention afforded an equivalent or greater reduction in the coefficient of friction when compared with conventional amide slip agents but the polymer plaques remained clear and transparent. In the examples of stearyl behenate, stearyl palmitate and ester formulation 4, which represent preferred embodiments of this invention, very low coefficients of friction were achieved (30% to 50% of the blank) at addition levels of only 0.2 to 0.3%.

It is of note that some esters of polyethylene glycol (not conforming to this invention) as exemplified by PEG 200 dierucate afforded an initial reduction in the coefficient of friction but this effect diminished over the 14-day test period. This renders compounds of this nature of little value as a long-term slip agents for PET.

.5

It is envisaged that mixtures of slip agents of general formula 1 can be used in polymers and co-polymers and that such agents may also be used in combination with known slip agents.

10 Thus it can be clearly seen that additives conforming to this invention provide the unique combination of a rapid and long-lasting reduction in the coefficient of friction of PET whilst maintaining low colour and high transparency.

15 Whilst it is known that certain compounds of general Formula 1 as defined herein have been reported as anti-slip agents in polymers this has generally not been reported in polyester-type polymers and in particular not in PET polymers as defined herein. The activity of certain compounds, including, but in no way limited to, the group comprising stearyl stearate, stearyl behenate, behenyl behenate, ethylene glycol distearate, ethyl behenate, behenyl acetate, palmityl myristate, 20 palmityl palmitate or mixtures thereof. This group of compounds provides slip values in the order of 40% or better at the test concentrations when compared to the blank. Such values are particularly high and represent a significant improvement on currently used additives in this context.

25 PREPARATION 1

Materials

85/90% behenic acid, AV=163.7 mg KOH/g, (mwt 342.7 g/mol), 200.0 g (0.584 30 mole).

n-butanol (butan-1-ol) 99.4+%(Aldrich), (mwt 74.12 g/mol), 400.0 g (5.397 mole).
sulphuric acid (98% min) catalyst, 4.0 g, or 1% on wt of butanol.

Procedure

A flask equipped with a stirrer and vertical water-cooled reflux condenser, was charged with the materials above and heated to and maintained at 110-120°C under
5 air at atmospheric pressure with constant reflux of butanol for approximately 4.5 hours. The resulting mixture was transferred to a separating funnel then 1 L of n-heptane @ 60°C added, followed by 2 L of saturated brine solution. The whole mixture was shaken and the phases allowed to separate. The aqueous phase was run off, and the heptane phase retained. The heptane phase was then repeatedly
10 washed with ca. 1 L of warm (60°C) distilled water until washings were pH 6. The heptane phase was then dried over anhydrous sodium sulphate and filtered. The filtrate was evaporated to constant weight on a rotary evaporator @ 70°C under 700 mmHg vacuum. Finally, the headspace of the vessel was blown with nitrogen for ca. 15 minutes to remove traces of butanol smell.

15

Yield 186g (theoretical maximum yield=232.9 g) 80% yield.

Analysis of final product:

AV	4.6	mg KOH/g
20 OHV	10.0	mg KOH/g
Sap value	137.3	mg KOH/g
moisture	0.02	% wt
colour	156	Hazen

Claims:

1. Use of a compound of general Formula 1 as a slip agent in a polyester polymer:

5

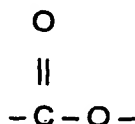


wherein: R and R¹ represent hydrocarbon moieties, each hydrocarbon moiety comprising 1 to 34 carbon atoms and wherein R and/or R¹ may be linear, branched chain, saturated or contain one or more double bonds and wherein; and wherein

10

X represents:

15



2. Use of a compound of general Formula 1 as a slip agent in a PET polymer as claimed in Claim 1 wherein the total number of carbon atoms in R, R¹ and X is greater than 16 and more preferably greater than 22.

20

3. Use of a composition of general Formula 1 as a slip agent as claimed in Claim 1 or Claim 2 wherein the total number of carbon atoms in R, R¹ and X is greater than 35.

25

4. Use of a composition of general Formula 1 as a slip agent in a polymer as claimed in any preceding claim wherein the total number of carbon atoms in R, R¹ and X is between 36 and 44.

5. Use of a composition of general Formula 1 as a slip agent in a PET polymer as claimed in any preceding claim wherein the composition of general Formula 1 is selected from the group comprising stearyl stearate, stearyl behenate and behenyl behenate, ethylene glycol distearate, ethyl behenate, behenyl acetate, palmityl myristate, palmityl palmate or mixtures thereof. This group of compounds provides slip values in the order of 40% or better at the test concentrations when compared to

35

the blank. Such values are particularly high and represent a significant improvement on currently used additives in this context.

6. Use of a composition of general Formula 1 as a slip agent in a polyester polymer as claimed in Claim 6 wherein the PET polymer is selected from the group comprising:-

poly(butylenes terephthalate)
 poly(cyclohexanedimethylene terephthalate)
 10 poly(ethylene isophthalate)
 poly(ethylene 2,6-naphthalenedicarboxylate)
 poly(ethylene phthalate)
 poly(ethylene terephthalate)
 and co-polymers thereof.

15

7. Use of a composition of general Formula 1 as a slip agent in a PET polymer according to any preceding claim wherein said composition of general Formula 1 is present in said PET polymer in an amount of between 0.1% to 1.0% wt/wt.

- 20 8. Use of a composition of general Formula 1 as a slip agent in a PET polymer according to Claim 7 wherein said composition is present in said PET polymer in an amount of between 0.2% to 0.75% wt/wt.

9. Use of a composition of general Formula 1 as a slip agent in a PET polymer
 25 substantially as herein described.

10. A polyester polymer incorporating one or more slip agents of general Formula 1:

30

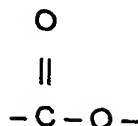


wherein: R and R¹ represent hydrocarbon moieties, each hydrocarbon moiety comprising 1 to 34 carbon atoms and wherein R and/or R¹ may be linear, branched chain, saturated or contain one or more double bonds; and wherein

35

X represents:

23



5

11. A polymer as claimed in Claim 10 incorporating one or more slip agents of general Formula 1 wherein the total number of carbon atoms in R, R¹ and X is greater than 16 and more preferably greater than 22.

10 12. A polymer as claimed in Claim 10 or Claim 11 wherein the total number of carbon atoms in R, R¹ and X is greater than 35.

13. A polymer as claimed in any of Claims 10, 11 or 12 wherein the total number of carbon atoms in R, R¹ and X is between 23 and 44.

15

14. A polymer as claimed in any of claims 10, 11, 12 or 13 incorporating one or more slip agents of general Formula 1 wherein the composition is selected from the group comprising stearyl stearate, stearyl behenate, behenyl behenate, ethylene glycol distearate, ethyl behenate, behenyl acetate, palmityl myristate, palmityl palmate or mixtures thereof.

20

15. A polymer as claimed in any of Claims 10 to 14 inclusive wherein said polymer is selected from a group comprising:-

25 poly(butylenes terephthalate)
poly(cyclohexanedimethylene terephthalate)
poly(ethylene isophthalate)
poly(ethylene 2,6-naphthalenedicarboxylate)
poly(ethylene phthalate)
30 poly(ethylene terephthalate)
or co-polymer thereof.

16. A polymer as claimed in any of Claims 10 to 15 inclusive incorporating one or more slip agents of general Formula 1 wherein said slip agent(s) are present in
35 said polymer in an amount of between 0.1% to 1.0% wt/wt.

17. A polymer as claimed in Claim 16 wherein said slip agent(s) are present in said polymer in an amount of between 0.2% to 0.75% wt/wt.
18. A polymer incorporating one or more slip agents of general Formula 1 substantially as herein described.
19. A method of treating a polyester polymer to increase the slip of said polymer said method comprising incorporating into said polymer a composition of general Formula 1 as defined in any of Claims 1 to 9 inclusive..
20. A method of treating a polymer as claimed in Claim 19 wherein said polymer is selected from a group comprising:-
- poly(butylenes terephthalate)
 - poly(cyclohexanedimethylene terephthalate)
 - poly(ethylene isophthalate)
 - poly(ethylene 2,6-naphthalenedicarboxylate)
 - poly(ethylene phthalate)
 - poly(ethylene terephthalate)
 - or co-polymer thereof.
21. A method according to Claim 20 wherein said composition of general Formula 1 is present in said polymer in an amount of between 0.1% to 1.0% wt/wt.
22. A method according to Claim 21 wherein said composition of general Formula 1 is present in said polymer in an amount of between 0.2% to 0.75% wt/wt.
23. A container made from a polymer as claimed in any of Claims 10 to 18 inclusive.
24. A container as claimed in Claim 23 wherein said container is formed from a polymer selected from a group comprising:-
- poly(butylenes terephthalate)
 - poly(cyclohexanedimethylene terephthalate)
 - poly(ethylene isophthalate)

25

poly(ethylene 2,6-naphthalenedicarboxylate)
poly(ethylene phthalate)
poly(ethylene terephthalate)
or co-polymer thereof.

5

25. A film made from a polymer as claimed in any of Claims 10 to 18 inclusive.

26. A film as claimed in Claim 25 wherein said film is formed from a polymer selected from a group comprising:-

10

poly(butylenes terephthalate)
poly(cyclohexanedimethylene terephthalate)
poly(ethylene isophthalate)
poly(ethylene 2,6-naphthalenedicarboxylate)

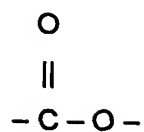
15

poly(ethylene phthalate)
poly(ethylene terephthalate)
or co-polymer thereof.

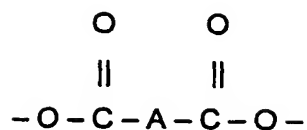
27. A composition comprising a copolymer of a polyester and a compound of general Formula 1 wherein: R and R¹ represent hydrocarbon moieties, each hydrocarbon moiety comprising 1 to 34 carbon atoms and R and/or R¹ may be linear, branched chain, saturated or contain one or more double bonds; and wherein X represents one of the moieties:

20

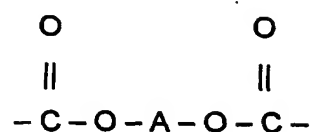
26



5



10



wherein A represents a hydrocarbon moiety comprising 2 to 36 carbon atoms and
 15 may be linear, branched chain, saturated or contain one or more double bonds.

1/1

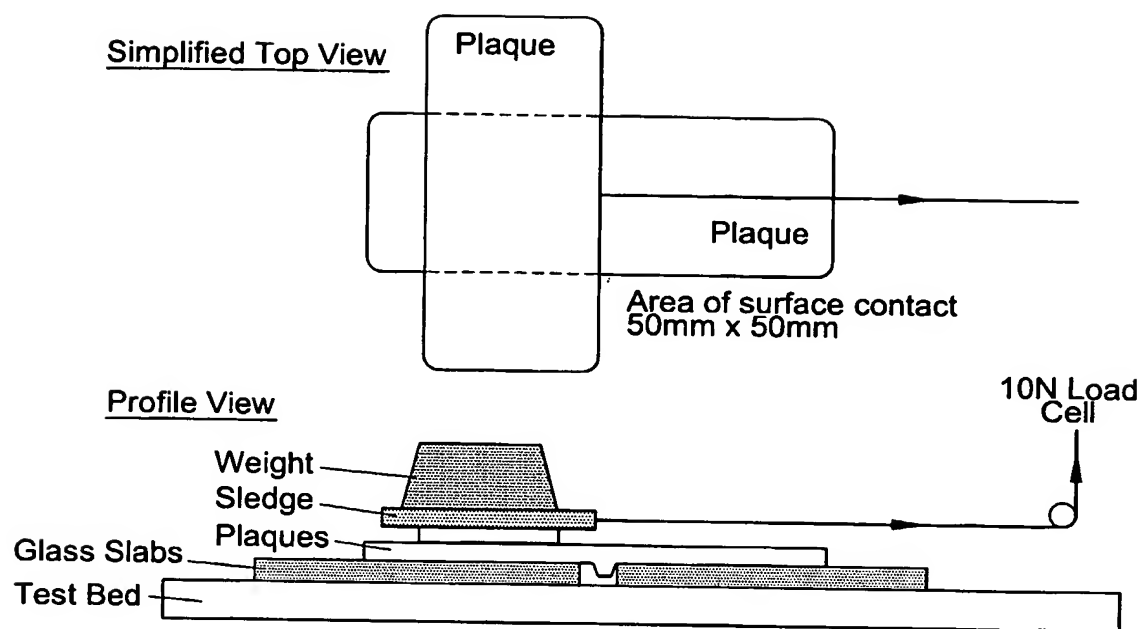


Fig. 1

INTERNATIONAL SEARCH REPORT

International Application No

PCT/GB2005/000805

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C08K5/101

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C08K

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, PAJ, WPI Data

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	PATENT ABSTRACTS OF JAPAN vol. 1998, no. 02, 30 January 1998 (1998-01-30) & JP 09 272191 A (KANEBO LTD), 21 October 1997 (1997-10-21) abstract paragraph '0022! paragraph '0039!; table 1	1-27
X	EP 0 947 543 A (SUMITOMO BAKELITE COMPANY LIMITED) 6 October 1999 (1999-10-06) claims 1,9 page 4; tables 1,2 ----- -/--	1-22, 25-27

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

* Special categories of cited documents:

- *A* document defining the general state of the art which is not considered to be of particular relevance
- *E* earlier document but published on or after the international filing date
- *L* document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- *O* document referring to an oral disclosure, use, exhibition or other means
- *P* document published prior to the international filing date but later than the priority date claimed

- *T* later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
- *X* document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
- *Y* document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.
- *G* document member of the same patent family

Date of the actual completion of the international search

21 July 2005

Date of mailing of the international search report

04/08/2005

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2
NL - 2280 HV Rijswijk
Tel (+31-70) 340-2040, Tx. 31 651 epo nl,
Fax: (+31-70) 340-3016

Authorized officer

Öhm, M

INTERNATIONAL SEARCH REPORT

International Application No
PCT/GB2005/000805

C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	<p>PATENT ABSTRACTS OF JAPAN vol. 2003, no. 12, 5 December 2003 (2003-12-05) & JP 2004 012610 A (TORAY IND INC), 15 January 2004 (2004-01-15) abstract paragraph '0004! paragraph '0016!</p>	<p>1-22, 25-27</p>
X	<p>US 6 326 435 B1 (TAKAYAMA KATSUNORI ET AL) 4 December 2001 (2001-12-04) claims 1,2,12 column 4, line 42 - line 45 column 8, line 20 - line 24 column 11 - column 16; tables 1-4</p>	<p>1-22,27</p>

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/GB2005/000805

Patent document cited in search report		Publication date	Patent family member(s)	Publication date
JP 09272191	A	21-10-1997	NONE	
EP 0947543	A	06-10-1999	CN 1236695 A ,C DE 69925665 D1 EP 0947543 A2 JP 11343353 A US 5998005 A	01-12-1999 14-07-2005 06-10-1999 14-12-1999 07-12-1999
JP 2004012610	A	15-01-2004	NONE	
US 6326435	B1	04-12-2001	WO 0055256 A1	21-09-2000